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Potential of sawdust materials for the removal of dyes and heavy metals: examination of isotherms and kinetics

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ABSTRACT

Sawdust is abundantly available from the timber and forest industry and has been studied in recent past as an adsorbent. This paper reviews the reported work on the uptake of dyes and heavy metals by natural and modified sawdust during the last 10 years. Research works examine the performance of isotherm models against the experimental equilibrium data, and an attempt has been made to discuss the kinetics of adsorption of metal ions and dyes on various sawdust materials on the basis of published report. Regeneration of sawdust materials has also been reviewed. It is found that pine, beech and mansonia sawdust are the most extensively studied adsorbents, whereas Pb^{2+} and methylene blue are the most efficiently removed pollutants, the Langmuir and Freundlich adsorption isotherms provide the best fit in most of the cases, and in general, pseudo-second-order kinetics is followed. There are very limited column studies and no report on commercial plant. Sawdust has a great potential in the wastewater treatment due to its abundant and cheap availability.

Keywords: Dyes; Heavy metal; Sawdust; Adsorption; Kinetic; Isotherm

1. Introduction

Heavy metals and dyes are often discharged by a number of industries, such as metal processing and dyeing-related industries. Metals can be distinguished from other toxic pollutants, since they are not biodegradable and wastewaters containing dyes are difficult to remove because of their inert properties. These unwanted chemicals cause health problems, when they exceed tolerance limit in water. Elimination of these toxic metal ions and dye ions from water and wastewater is therefore very important and urgent action needs to be taken to protect both public health and the environment. The removal of heavy metals and dyes from wastewater can be achieved by several techniques such as ion exchange, precipitation, coagulation, reverse osmosis, membrane separation, electrolysis [1–3], aerobic and anaerobic treatment [4], bacterial treatment [5], electrodialysis [6], magnetic separation [7], microbial reduction [8], photochemical reactions [9], ultrasonic treatment [10] and adsorption [11]. Among these, adsorption is the most used process for its inexpensiveness, ease of operation and insensitivity to toxic pollutants [12,13]. The activated carbon is a commercial adsorbent for eliminating pollutions from wastewater. However, the higher

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production cost, and regeneration difficulty of activated carbon limit its widespread use [14]. Thus, the development of cheaper, eco-friendly and more efficient adsorbent is a subject of intensive research. Most of the adsorbate species are adsorbed onto the interface within a short interval of contact time [15–17]. The reason for this short contact time is that a two-step adsorption mechanism occurs which involves a rapid metal uptake taking place through external and internal diffusion during the first step, and intraparticle diffusion controls the adsorption rate during the second step [18]. However, a longer contact time, often on the order of a few hours, is often needed for the attainment of equilibrium [19–22]. Indeed, later, due to the decrease in the number of sorption sites on the adsorbent as well as adsorbate concentration, the sorption of adsorbate became slow. In addition, sorption efficiency and costs are rather variable. In spite of these problems, opportunities for finding new sorbents and new areas of sorbent application are open. Various adsorbents have been used to remove different types of heavy metal and dye ions from wastewater especially those that are harmful to mankind. Sawdust is actually an efficient adsorbent that is effective to many types of inorganic and organic pollutants [23–29]. Hence, the utilization of such waste is most desirable. Therefore, the aim of this study was to outline the current applications of sawdust in dyes and heavy metals adsorption and to review recent information concerning the factors that impact the adsorption, the system equilibrium and kinetics with respect to various models that have been applied in the literature.

2. Sawdust as an adsorbent for water treatment

Sawdust is a waste by-product of the timber industry and is an abundant residue from the forest industry. Sawdust materials are biodegradable and have an affinity for water. Wood powder does not noticeably swell in water and does not decompose upon prolonged contact with water [30]. Sawdust can be used as a low-cost adsorbent due to its lignocellulosic composition [31–33]. It contains abundant lignin, cellulose, hemicellulose and some functional groups such as hydroxyl, carboxyl, amide and phenolic groups in its structure, which make the adsorption processes possible. These polar functional can be involved in chemical adsorption phenomenon [32,34]. Metal and dye ions' adsorption is associated with the existence of an adsorbent surface of ligands like carboxyl, amide and hydroxyl groups, which have a different affinity for metal and dye ions binding. Their presence in the solution can lead to the formation of various types of interactions that may affect the affinity and adsorption capacity. One problem with sawdust materials is that sawdust contains tannins in low concentration usually mentioned as toxic constituent. We must prevent the elution of tannin compounds that could stain the treated water [35]. Modification of sawdust using physical or chemical methods can be performed with the aim of enhancing the capability and efficiency of sawdust adsorption. Zou et al. [32] modified *Pinus tabulaeformis* sawdust with citric acid for methylene blue adsorption. After modification, sawdust possesses more carboxyl groups. The carboxyl group is turned into –COO−, a significantly high electrostatic attraction exists between the negatively charged surface of adsorbent and cationic methylene blue, leading to maximum dye adsorption. Pereira et al. [36] modified *manilkara* sawdust with ethylene diamine tetra acetic dianhydride (EDTAD) in order to introduce carboxylic acid and amine functional groups into this material. Zn(II) ions were adsorbed onto the sorbent by ion exchange and surface adsorption mechanisms. Phosphoric acid was used for modification of *oak* sawdust for lead removal [37]. After modification, a seven-fold increase in sorption capacity for Pb(II) was observed. An enhanced adsorption capacity as a result of surface modification was also observed by other research groups [38–41]. A scanning electron microscope (SEM) was used to observe the surface structure of aleppo *pine* tree sawdust [42]. Many cavities of various dimensions were clearly evident on the surface of aleppo *pine* tree sawdust. SEM picture indicated texture and large porosity. These properties promise sawdust a variety of current applications in pollutant removal from aqueous solutions [43–53]. If sawdust could be used as adsorbent, both the environment protection and wooden industry could benefit.

2.1. Adsorption isotherms

Models have an important role in technology transfer from a laboratory to industrial scale. Appropriate models can help in understanding process mechanisms, analyse experimental data, predict answers to operational conditions and optimize processes. The relationship, at given temperature, between the equilibrium amount of the pollutants adsorbed and those remaining in solution is called the adsorption isotherm [54].

Out of the several possible isotherms, the Langmuir and Freundlich isotherm models have been used successfully by several researchers to describe the equilibrium sorption data between the pollutant in the aqueous phase and solid (sawdust material) phase.

The Langmuir model was derived on the basis of three assumptions and may be written in the form:

$$q_e = q_{max}\left(\frac{b\,C_e}{1 + b\,C_e}\right) \quad (1)$$

where q_{max} is the maximum amount of adsorbate adsorbed by the adsorbent to achieve complete monolayer coverage of the adsorbent surface (mg/g) and b is the Langmuir affinity constant (l/mg). The variables q_e (mg/g) and C_e (mg/l) are the amounts of pollutant adsorbed per unit weight of adsorbent and the equilibrium concentration, respectively.

The parameter q_{max} is assumed to correspond to saturation of a fixed number of identical surface sites and b is related to the adsorption energy.

The isotherm developed by Freundlich describes the equilibrium on heterogeneous surfaces and does not assume monolayer capacity. The Freundlich equation is:

$$q_e = K_F\,C_e^{\frac{1}{n}} \quad (2)$$

K_F is a constant indicative of the adsorption capacity of the adsorbent ((mg/g)(l/mg)$^{1/n}$) and n is an empirical constant related to the magnitude of the adsorption driving force.

2.1.1. Adsorption of dyes

The industry emits dyes which are the indicator of water pollution, into wastewater. Many dyes are harmful to human beings and have considerable resistance to biodegradation. This section is focusing on the isotherm modelling of dyes adsorption by sawdust.

Zou et al. [32] used citric acid-modified pine sawdust for removing methylene blue by batch method. They studied four types of isotherms which are Freundlich, Redlich–Peterson, Dubinin–Radushkevich and Langmuir with the latter fitting the experimental data better than the other isotherms.

Zhang et al. [41] also used citric acid-modified *pine* sawdust for removing methylene blue. Generalized isotherm, Langmuir and Freundlich isotherms were used to describe the adsorption experiment in single system. Both Generalized isotherm and Langmuir isotherm equations could fit the equilibrium data. Maximum removal of methylene blue was 76.32 mg/g.

Megat Hanafiah et al. [55] investigated batch sorption of aqueous acid blue 25 using pretreated *Shorea dasyphylla* sawdust. Adsorption isotherms were modelled with Langmuir and Freundlich equations. The adsorption data fit the Langmuir model well. The maximum adsorption capacity for acid blue 25 at pH 2 was 24.39 mg/g.

Ouazene and Lounis [56] reported that one gram of Aleppo pine tree sawdust could remove 91.5% of the basic blue 3 from the solution of 30 mg/l (20 ml) at pH 7 with a shaking time of 300 min. This adsorbent holds a relatively high maximum adsorption capacity (65 mg/g).

Khattri and Singh [57] utilized *sagaun* sawdust for crystal violet removal. Isotherm data best fitted the Langmuir isotherm with maximum adsorption capacity of 4.26 mg/g at pH 7.5 and 25°C temperature.

Aleppo pine tree sawdust was utilized for the removal of astrazon yellow from aqueous solution [58]. The adsorption data fit the Langmuir model well. The maximum astrazon yellow uptake was 81.77 mg/g at pH 7.5.

Gong et al. [59] studied adsorption of malachite green using native and modified *Castanopsis sclerophylla* sawdust. The data fitted well the Langmuir isotherm. Maximum adsorption capacities of native and modified sawdust were 85.47 and 196.08 mg/g, respectively.

Ansari and Mosayebzadeh [60] studied adsorption of methylene blue using native and treated *walnut* sawdust in batch and column studies. Both Langmuir and Freundlich isotherm equations could fit the batch equilibrium data. Maximum adsorption capacities of native and treated sawdust were 19.41 and 34.36 mg/g, respectively. The breakthrough curves obtained at different flow rates in column studies revealed that treated sawdust is an excellent adsorbent for the removal of methylene blue [60].

Sawdust was modified with ethylenediamine for the removal of methyl violet from water [61]. Equilibrium time for methyl violet removal was 1 h. Solution pH significantly affected the adsorption, and the adsorption capacity increased with the increase in pH. The Langmuir isotherm was the best-fit adsorption isotherm model for the experimental data. Adsorption capacity of modified sawdust was found to be 100.20, 131.58 and 161.29 mg/g at temperature 20, 40 and 50, respectively.

Jain et al. [62] investigated batch sorption of aqueous basic swiss blue dye using pretreated *Acacia nilotica* sawdust. Sawdust was subjected to formaldehyde pretreatment (FSD) and boiling (BSD). The data followed Langmuir model. Maximum adsorption capacities of formaldehyde-treated sawdust (FSD) and boiled sawdust (BSD) were found to be 45.66 and 41.84 mg/g of biomass, respectively, at pH 6.

Witek-Krowiak [63] reported batch sorption of aqueous malachite green using *beech* sawdust. The biosorption equilibrium data were correlated by Langmuir, Freundlich, Temkin, Redlich–Peterson and Sips isotherms. Redlich–Peterson model correlated best with equilibrium data. The maximum adsorption capacity of beech sawdust was 83.21 mg/g at pH 5 and 20°C.

Dulman and Cucu-Man [64] applied Langmuir model to adsorption data of direct brown, direct brown 2 and basic blue 86 by *beech* wood sawdust. Maximum removal of direct brown and direct brown 2 were 526.3 and 416.7 mg/g, respectively, at pH 3 and for basic blue 86, 136.9 mg/g at pH 7.

Meranti tree is a common tree present in all tropical countries such as Malaysia and Indonesia. The ability of *meranti* sawdust to remove methylene blue from aqueous solutions was evaluated by Ahmad et al. [65]. Results of this study show that pH 9 is favourable for the adsorption of methylene blue. The isothermal data could be well described by the Langmuir equation. The ultimate capacity obtained was 120.48 mg/g.

Sidiras et al. [66] reported that the adsorption of methylene blue onto natural and autohydrolysed *pine* sawdust fitted to Sips model, while bismarck brown and acridine orange fitted to Freundlich isotherm [66]. Adsorption capacities are reported in Table 1. Bohart–Adams model and Clark model were applied to column studies and parameters necessary for the design of a packed bed reactor were evaluated. The Bohart–Adams represents well the sorption of three dyes used.

Bello et al. [67] utilized *Gmelina arbhoria* sawdust for aqueous methylene blue removal in batch experiments. The data were in good agreement with Langmuir model. The monolayer adsorption capacity was evaluated as 263.16 mg/g at pH 7.

Khattri and Singh [68] reported batch sorption of malachite green using *neem* sawdust. The adsorption of malachite green is well described by Langmuir isotherm, which expresses the existence of monolayer adsorption.

The isotherm studies for malachite green adsorption by *rattan* sawdust conducted by Hameed and El-Khaiary [69] showed that the Langmuir isotherm fitted well with the experimental data. A maximum sorption capacity of 62.71 mg/g was achieved.

Pekkuz et al. [70] studied batch removal of metanil yellow and methylene blue using *poplar* sawdust. Maximum removal of methylene blue and metanil yellow were 5.74 and 1.34 mg/g, respectively.

Mansonia sawdust adsorbed methylene blue and methyl violet from aqueous solutions by batch method [71]. Author reported the adsorption capacity of methylene blue and methyl violet to be 28.89 and 16.11 mg/g, respectively, at pH 10.

The isotherm studies for methyl violet adsorption by *mansonia* sawdust conducted by Ofomaja and Ho [72] showed that maximum methyl violet uptake was 24.1 mg/g at pH 7.

2.1.2. Adsorption of heavy metals

Heavy metals are toxic and harmful even at low concentrations. They are not biodegradable and tend to accumulate in living organisms, causing various diseases and disorders [73,74].

Bozic et al. [34] performed a series of batch experiments for the removal of three heavy metals, i.e. Cu (II), Ni(II) and Zn(II) by *beech* sawdust. The data followed Langmuir model.

Pereira et al. [36] explored the use of modified *manilkara* sawdust for aqueous Zn(II) removal. Adsorption data fitted well to Langmuir isotherm and the maximum adsorption capacity was 80 mg/g.

Jeon and Kim [37] examined lead adsorption by modified *oak* sawdust. The sorption of Pb(II) is well described by the Langmuir isotherm. The maximum adsorption capacity was 300.44 mg/g of biomass at pH 4.

Rehman et al. [38] evaluated the sorption potential of *Dalbergia sissoo* sawdust for the removal of nickel ions. Both Langmuir and Freundlich isotherm equations could fit the equilibrium data.

Zhang et al. [41] investigated modified *pine* sawdust for removing methylene blue and copper in single and binary compound systems. Both Generalized isotherm and Langmuir isotherm models were suitable for describing the adsorption of Cu(II) by *pine* sawdust. Maximum removal of copper was 13.72 mg/g at pH 5.

Bozic et al. [75] removed Cu(II), Cd(II), Zn(II), Mn(II) and Ni(II) metal ions by *linden* sawdust from aqueous solution. The data followed Langmuir model, and *linden* sawdust adsorption capacities are reported in Table 2.

Cu(II) was removed from *maple* wood sawdust by batch method [76]. A maximum adsorption of 9.51 mg/g Cu(II) was achieved at pH 6.

Witek-Krowiak [77] used *beech* sawdust for removal of Cu(II) and Cr(III) ions from waters. Maximum removal of Cu(II) and Cr(III) were 30.22 and 41.86 mg/g, respectively, at pH 5.

Ahmad et al. [78] used *D. sissoo* sawdust modified with HCl for adsorption of Zn(II) and Cd(II). The data followed Langmuir model. Maximum removal of Zn (II) and Cd(II) were 49.57 mg/g (pH 10) and 95.31 mg/g (pH 6.7), respectively.

Table 1
Results from selected dye adsorption by sawdust materials

Sawdust source	Dye	pH	Equilibrium model	Adsorption capacity (mg/g)	Refs.
A. nilotica	Swiss blue dye	6	Langmuir	41.84	[62]
A. nilotica	Swiss blue dye	6	Langmuir	45.66	[62]
Aleppo pine tree	Basic Blue 3	7–11	Langmuir	65.36	[56]
Aleppo pine tree	Astrazon Yellow	7.5	Langmuir	81.77	[58]
Beech	Malachite Green	5	Redlich–Peterson	83.21	[63]
Beech	Direct Brown	3	Langmuir	526.3	[64]
Beech	Direct Brown 2	3	Langmuir	416.7	[64]
Beech	Basic Blue 86	7	Langmuir	136.9	[64]
C. camphora	Malachite Green	7	Langmuir	155	[118]
C. sclerophylla	Malachite Green	8	Langmuir	85.47	[59]
Gmelina arbhoria	Methylene Blue	7	Langmuir	263.16	[67]
Mansonia	Methylene Blue	10	Langmuir	28.89	[71]
Mansonia	Methyl Violet	10	Langmuir	16.11	[71]
Mansonia	Methyl Violet	7	Langmuir	24.1	[72]
Meranti	Methylene Blue	9	Langmuir	120.48	[65]
Neem	Malachite Green	7.2	Langmuir	4.35	[68]
Pine	Methylene Blue	5	Langmuir	75.33	[41]
Pine	Methylene Blue	8	Sips	40.42	[66]
Pine	Bismarck Brown	8	Freundlich	3.37	[66]
Pine	Acridine Orange	8	Freundlich	3.11	[66]
Pine	Methylene Blue	8	Sips	138.1	[66]
Pine	Bismarck Brown	8	Freundlich	15.20	[66]
Pine	Acridine Orange	8	Freundlich	15.52	[66]
Pinus silvestris	Malachite Green	5	Sips	71.67	[94]
Pinus tabulaeformis	Methylene Blue	6.5	Langmuir	111.45	[32]
Poplar	Metanil Yellow	6.23	Langmuir	1.34	[70]
Poplar	Methylene Blue	6.38	Langmuir	5.74	[70]
Rattan	Malachite Green	10	Langmuir	62.71	[69]
Sagaun	Crystal Violet	7.5	Langmuir	4.26	[57]
Shorea dasyphylla	Acid Blue 25	2	Langmuir	24.39	[55]
Walnut	Methylene Blue	6	Langmuir	19.41	[60]
Walnut	Methylene Blue	12	Langmuir	34.36	[60]

Zakaria et al. [79] reported batch sorption of aqueous Cr(VI) using untreated *rubber* wood sawdust. The data fitted well the Langmuir isotherm. The maximum adsorption capacity was 4.87 mg/g of biomass at pH 2.

The adsorptions of lead from aqueous solutions by Brazilian sawdust samples (*Caryocar* spp., *Manilkara* spp. and *Tabebuia* spp.) were investigated by Prado et al. [80]. Maximum adsorption capacities of *Caryocar* spp., *Manilkara* spp. and *Tabebuia* spp. were 89.10, 145.04 and 95.31 mg Pb(II)/g of dry adsorbents, respectively.

Musapatika et al. [81] studied batch Co(II) removal by *pine* sawdust using a 2^3 factorial experimental design. Equilibrium data fitted well to Freundlich isotherm model, which suggested that adsorption occurred on the heterogeneous surface.

Sidiras et al. [82] studied adsorption capacity of *Pinus sylvestris* sawdust after autohydrolysis treatment for the removal of Cr(VI) metal ions. The isothermal data could be well described by the Freundlich equation. Adsorption capacities of native and modified sawdust were found to be 2.28 and 8.93 mg/g, respectively, at pH 2.

Kapur and Mondal [83] removed Cr(VI) from aqueous solution using *Mangifera indica* sawdust by batch mode. A maximum sorption capacity of 10.86 mg/g was achieved at pH 2.

Mishra et al. [84] explored the use of *Cedrus deodara* sawdust for aqueous Zn(II) removal in batch experiments. The ultimate capacity obtained from the Langmuir model was 97.39 mg/g at pH 5.

Andrabi [85] used *Cordia africana* sawdust for the removal of lead and nickel from aqueous solutions.

Table 2
Results from selected heavy metal adsorption

Sawdust source	Metal	pH	Equilibrium model	Adsorption capacity (mg/g)	Refs.
Acacia Arabica	Cr(VI)	6	Langmuir	111.61	[90]
Acacia Arabica	Pb(II)	6	Langmuir	52.38	[90]
Acacia Arabica	Hg(II)	6	Langmuir	20.62	[90]
Acacia Arabica	Cu(II)	6	Langmuir	5.64	[90]
Beech	Cu(II)	>4	Langmuir	4.5	[34]
Beech	Ni(II)	>4	Langmuir	4	[34]
Beech	Zn(II)	>4	Langmuir	2	[34]
Beech	Cu(II)	5	Langmuir	30.22	[77]
Beech	Cr(III)	5	Langmuir	41.86	[77]
Birch	Cu(II)	5.5	Langmuir	4.90	[97]
Black locust	Cu(II)	4	Langmuir	4.38	[91]
Black locust	Zn(II)	6	Langmuir	5.16	[91]
Caryocar spp.	Pb(II)	5	Langmuir	89.10	[80]
Cedrus deodara	Zn(II)	5	Langmuir	97.39	[84]
Cordia Africana	Pb(II)	7	Langmuir	211.34	[85]
Cordia Africana	Ni(II)	7	Langmuir	123.27	[85]
Dalbergia sissoo	Ni(II)	6	Langmuir	10.47	[38]
Dalbergia sissoo	Zn(II)	10	Langmuir	49.57	[78]
Dalbergia sissoo	Cd(II)	6.7	Langmuir	95.31	[78]
Linden	Cd(II)	3.5–5.5	Langmuir	3.5	[75]
Linden	Zn(II)	3.5–5.5	Langmuir	2.17	[75]
Linden	Ni(II)	3.5–5.5	Langmuir	4.6	[75]
Linden	Mn(II)	3.5–5.5	Langmuir	1	[75]
Mangifera indica	Cr(VI)	2	Langmuir	10.86	[83]
Manilkara spp.	Pb(II)	5	Langmuir	145.05	[80]
Manilkara spp.	Zn(II)	6.3	Langmuir	80	[36]
Mansonia	Cu(II)	6	Freundlich	42.37	[87]
Mansonia	Pb(II)	6	Freundlich	51.81	[87]
Maple	Cu(II)	6	Langmuir	9.19	[76]
Meranti	Cu(II)	6	Langmuir	32.05	[86]
Meranti	Cr(III)	6	Langmuir	37.88	[86]
Meranti	Ni(II)	6	Langmuir	35.97	[86]
Meranti	Pb(II)	6	Langmuir	34.25	[86]
Oak	Pb(II)	4	Langmuir	300.44	[37]
Pine	Cu(II)	1.5	Langmuir	24.3	[89]
Pine	Cu(II)	5	Langmuir	13.72	[41]
Pinus sylvestris	Cr(VI)	2	Freundlich	8.93	[82]
Pinus halepensis	Cd(II)	9	Freundlich	5.36	[88]
Poplar	Zn(II)	4	Langmuir	0.97	[73]
Poplar	Cd(II)	4	Langmuir	0.16	[73]
Quercus coccifera	Cu(II)	4	Langmuir	3.22	[98]
Quercus coccifera	Ni(II)	8	Langmuir	3.29	[98]
Quercus coccifera	Cr(VI)	3	Langmuir	1.70	[98]
Rubber	Cr(VI)	2	Langmuir	4.87	[79]
Tabebuia spp	Pb(II)	5	Langmuir	95.31	[80]

Both Langmuir and Freundlich isotherm equations could fit the equilibrium data. Maximum removal of Pb(II) and Ni(II) were 211.34 and 123.27 mg/g, respectively, at pH 7.

Rafatullah et al. [86] removed Cu(II), Cr(III), Ni(II) and Pb(II) metal ions using *meranti* sawdust. The adsorption equilibrium data were correlated by Langmuir, Freundlich and Dubinin–Radushkevich (D–R) isotherms. The data followed Langmuir, and maximum removal of Cu(II), Cr(III), Ni(II), Pb (II) were 32.05, 37.88, 35.97, 34.25, respectively, at pH 6.

Ofomaja et al. [87] used *mansonia* sawdust for the removal of Cu(II) and Pb(II) from aqueous solution. The Langmuir isotherm provided a better fit to experimental data for lead ion sorption, while copper ion sorption was best described by the Freundlich and BET isotherms. *Mansonia* sawdust adsorption capacities are reported in Table 2.

Semerjian [88] examined batch cadmium adsorption by untreated *Pinus halepensis* sawdust. The data followed Freundlich model with maximum uptake 5.36 mg/g at pH 9.

The adsorption of copper ions onto *pine* sawdust was reported by Hansen et al. [89]. The sorption data fitted to both Langmuir and Freundlich isotherms. The ultimate capacity obtained from the Langmuir model was 24.3 mg/g at pH 1.5.

Modified *Acacia arabica* sawdust was tried as an adsorbent for the removal of Cr(VI), Pb(II), Hg(II), Cu (II)) metal ions by Meena et al. [90]. Both Langmuir and Freundlich isotherm equations fitted adsorption well, which indicated adsorption by combined mechanisms onto a heterogeneous surface. Sciban et al. [91] evaluated the sorption potential of modified *hardwood* sawdust for the removal of copper ions and zinc ions. Both Langmuir and Freundlich isotherm equations could fit the equilibrium data

Stankovic et al. [92] used *linden* and *poplar* sawdust for the removal of copper from water. Both batch and column tests were carried out. It was found that *poplar* and *linden* sawdust have both almost equal adsorption capacities against copper ions. Higher sorption capacities were obtained from the column experiments than from the batch tests.

2.2. Adsorption kinetics

Kinetics studies give information on sorbate uptake rates and rate controlling steps such as external mass transfer, intraparticle mass transfer and adsorption process [93].

The pseudo-first-order equation is generally expressed as follows:

$$\log(q_e - q_t) = \log q_e - \frac{k_1}{2.303} t \quad (3)$$

where q_e and q_t are the values of amount adsorbed per unit mass at equilibrium and at any time t, k_1 is the first-order rate constant in s^{-1}.

If the rate of sorption is a second order mechanism, the pseudo-second-order kinetic rate equation is expressed as:

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \quad (4)$$

where k_2 is second order rate constant in (g/mg/min).

The rate, q_t, for intraparticle diffusion k_{int} is:

$$q_t = k_{int} t^{0.5} \quad (5)$$

where k_{int} is the intraparticle diffusion rate constant.

2.2.1. Kinetic modelling of dyes

Zou et al. [32] reported that the adsorption of methylene blue onto modified sawdust (*P. tabulaeformis*) followed the Elovich model and the process involving both boundary layer and intra-particle diffusion processes [32].

Zhang et al. [41] found that adsorption of methylene blue and copper onto citric acid-modified *pine* sawdust was described by pseudo-second-order kinetic model from the single and binary compound systems.

Megat Hanafiah et al. [55] computed kinetic parameters for the adsorption of acid blue 25 onto *S. dasyphylla* sawdust in a batch system. The acid blue 25 sorption kinetics were described by a pseudo-second-order kinetic model.

Ouazene and Lounis [56] reported the kinetics of basic blue 3 onto Aleppo pine-tree sawdust. It was found that both external mass transfer and intraparticle diffusion played the chief role in the adsorption mechanisms of dye, and adsorption kinetics followed the pseudo-second-order kinetic model.

Khatri and Singh [57] reported that rate of adsorption of crystal violet ions onto *sagaun* sawdust was very high initially and maximum adsorption occurs within 14 min. The data followed a pseudo-first-order kinetic model.

Sahmoune and Ouazene [58] proposed an adsorption mechanism and reported a consecutives steps shown in Fig. 1 for the removal of astrazon yellow (AY) by Aleppo pine-tree sawdust. The intraparticle diffusion coefficient and external mass transfer were 4.2 10^{-11} cm^2/s and 8.5 10^{-5} m^{-1}, respectively, at initial concentration 50 mg/l of astrazon yellow. AY sorption onto sawdust was mainly located on the surface.

Gong et al. [59] reported malachite green adsorption native and modified *C. sclerophylla* sawdust in batch experiments. Their kinetic results best fitted the pseudo-second-order model, indicating that the rate determining step involved chemisorption.

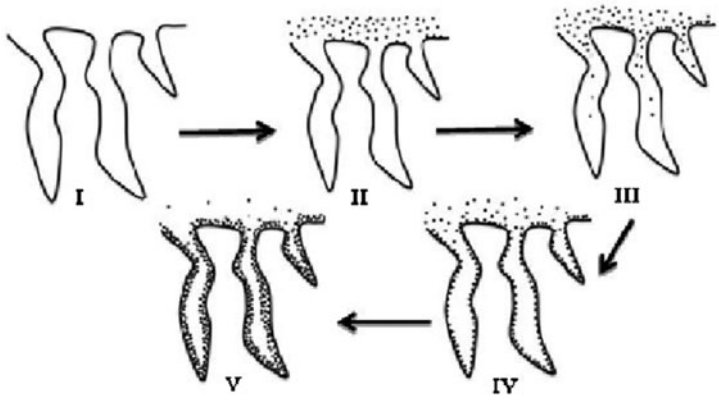

Fig. 1. Mass-transfer processes in the adsorption of cationic dye by sawdust, (I): porous adsorbent, (II): dye transfer from the boundary film to the surface of the adsorbent, (III): transfer of the dye from the surface to the intraparticle actives sites, (IV) and (V): Uptake of dye ion on the active sites (monolayer and multilayer adsorption).

Work carried out by Witek-Krowiak [63] indicate that kinetics of malachite green biosorption onto *beech* sawdust parameters (time, concentration of the dye and biosorbent, pH, temperature, ionic strength, anionic surfactant presence) influence process kinetics of malachite green biosorption onto beech sawdust. Pseudo-second-order and general rate law model describe experimental points very well which corresponds to a chemisorption process. Also intra-particle diffusion plays a significant role in the adsorption mechanism.

Ahmad et al. [65] reported that the adsorption of methylene blue onto *meranti* sawdust followed the pseudo-first-order adsorption kinetics and the intraparticle diffusion is not the sole rate-limiting factor for the adsorption of methylene blue [65].

Hameed and El-Khaiary [69] reported the kinetics of malachite green onto *rattan* sawdust. The pseudo-first-order model fits the sorption data with high correlation coefficients. It was found that for a short time period (40 min), the rate of adsorption is controlled by film diffusion. However, at longer adsorption times, pore-diffusion controls the rate of adsorption.

Pekkuz et al. [70] analysed the adsorption of metanil yellow and methylene blue by *poplar* sawdust using pseudo-first-order and second-order kinetic models. The experimental data were observed to fit well to the pseudo-second-order equation. The correlation coefficient (R^2) was observed to be close to 1.c

Ofomaja [71] studied kinetics of adsorption of two dyes onto *mansonia* wood sawdust in a batch reactor using the pseudo-first-order equation and the pseudo-second-order equation. Adsorption kinetics followed the pseudo-second-order kinetic model. The process mechanism was found to be complex, consisting of both mass transfer and intraparticle diffusion. External mass transfer was claimed as the dominant mechanism.

Pinus silvestris sawdust was reported as a biosorbent for the removal malachite green from aqueous solution [94]. The biosorption kinetics followed the pseudo-second-order, which confirms the chemical nature of the rate-controlling elementary process.

Carra sawdust pre-treated with formaldehyde was used to adsorb reactive red 239 [95]. The kinetic data followed pseudo-second-order model. The mechanism of dye adsorption was a combination of external mass transfer and intra-particle diffusion.

Crystal violet and rhodamine B biosorption from aqueous solutions by sawdust was investigated by Parab et al. [96]. More than 80% adsorption was achieved within first 60 min for the removal of the two dyes. The application of various kinetic models reveals good correlation of a pseudo-second-order kinetics model with the experimental data.

2.2.2. Kinetic modelling of heavy metals

Ofomaja [33] studying the removal of copper ions from aqueous solution using untreated *mansonia* sawdust observed that although the pseudo-second-order model described the biosorption process for the whole biosorption period, intraparticle process was active in the rate-determining step especially in the second stage of the biosorption process.

Bozic et al. [34,75] reported the kinetics of heavy metals onto *beech* and *linden* sawdust. The overall adsorption process was best described by the pseudo-second-order kinetic model. The adsorption kinetics is fast and after 10–20 min of contact time, the system is close to equilibrium.

Witek-Krowiak [77] from her work on adsorption of Cu(II) and Cr(III) on *beech* sawdust has concluded that the process follow pseudo-second-order kinetics. For initial Cu(II) concentration range of 20–200 mg/l the rate coefficient decreased from 2.738 g/mg/min to 0.024 g/mg/min and also in adsorption of Cr(III) from aqueous solution on beech sawdust the rate coefficient decreased with increasing solute concentration. The adsorption was rapid in the first 20 min for both Cu(II) and Cr(III).

The adsorption of Cr(VI) on *Mangifera indica* sawdust had followed pseudo-first-order kinetics [83]. Nearly 30–40% of the adsorption capacity was realized within the first 10 min of contact and the adsorption equilibrium was established within short period of 60 min. Film-diffusion was found as the rate-limiting step for Cr(VI) adsorption from bulk of the liquid to the surface of the adsorbent.

Rafatullah et al. [86] used *meranti* sawdust for the uptake of Cu(II), Cr(III), Ni(II) and Pb(II) from aqueous solutions where the metal–sawdust interactions followed pseudo-second-order model. The rate of controlling step was mainly intraparticle diffusion but was not the only rate-limiting step for the metal ions [86].

Semerjian [88] reported the kinetic of cadmium ion onto untreated *P. halepensis* sawdust. The experimental data were observed to fit well to the pseudo-second-order equation. Author observed that intra-particle diffusion plots did not have linearity over the whole range of contact time, but linearity was maintained over a short time interval [88].

The pseudo-second-order model showed the best fit for kinetics of Cu(II) sorption by *pine* sawdust [89].

Meena et al. [90] reported that the biosorption of heavy metals onto *A. arabica* sawdust followed the pseudo-first-order adsorption kinetics.

Adsorption of copper onto *birch* sawdust was shown to follow pseudo-second-order kinetic model [97]. Argun et al. [98] analysed the biosorption of *Quercus coccifera* sawdust using pseudo-first-order and second-order kinetic models. The experimental data were observed to fit well to the pseudo-second-order equation. It was also shown that external mass transfer might also play some role.

The pseudo-second-order model assumes that two reactions are occurring, the first one is fast and reaches equilibrium quickly and the second is a slower reaction that can continue for long time periods [99]. The pseudo-second-order equation has also been interpreted as a special kind of Langmuir kinetics [100]. This line of interpretation assumes that: (i) the adsorbate concentration is constant in time and (ii) the total number of binding sites depends on the amount of adsorbate adsorbed at equilibrium [100].

Li et al. [101] studied the biosorption of lead(II), chromium(III) and copper(II) by sawdust and modified peanut husk, the authors concluded that intra-particle diffusion was not rate-controlling step. But rather the pseudo-second-order model pseudo-second-order model described the biosorption over the whole range of contact time.

3. Influence of adsorption conditions on the removal of dyes and heavy metals by sawdust

Factors such as pH, temperature, ionic strength, etc., have various degrees of influence on the pollutant uptake by the sawdust material. This section is intended to give a brief discussion on these parameters.

3.1. Influence of pH

The initial pH of solution can significantly influence adsorption of dyes and metals [102,103]. It determines the surface charge of the adsorbent and the degree of ionization and speciation of the adsorbate. So, it is very important to consider the ionic states of the functional groups of the adsorbent as well as the metal solution chemistry at different pH values. The influence of metal speciation in solutions certainly requires attention. Metal ions are precipitated out in alkaline pH range and, the chemical speciation of metal is decided by solution pH [104,105].

The pH is a significant factor for determining the existing form of the metallic species in aqueous solution. For example, It is known that lead species are mainly present in the forms of Pb^{2+}, $Pb(OH)^{+}$ and $Pb(OH)_2$ at $pH < 9$ [106]. At $pH < 6$, the predominant lead species is Pb^{2+}. Pb^{2+} adsorption that took place at low pH can be attributed mainly to the competition between H^{+} and Pb^{2+} ions on the surface sites [104,105]. At pH 6–7, the main lead species are Pb^{2+} and $Pb(OH)^{+}$, and the electrostatic attraction between the negatively charged sawdust surface and the positively charged Pb^{2+} and $Pb(OH)^{+}$ may be responsible for the removal of lead(II) [106].

In general, as solution pH increases, most cationic dye sorption or cationic metal sorption is enhanced with pH, increasing to a certain value followed by a reduction on further pH increase. The opposite trend happens for anionic dye and anionic metal sorption. At low pH, competition occurs between protons and metal cations, leading to less metal uptake. At higher

pH values, the metal sorption stops and the hydroxide precipitation starts [34,75].

The pH at which the surface charge is zero is called the point of zero charge (pzc). Many researchers studied the zero point charge (pHzpc) of the adsorbent that was prepared from sawdust materials in order to better understand the adsorption mechanism. The surface charge of the adsorbent is positive when the media pH is below the pHzpc value, while it is negative at a pH over the pHzpc. Cationic dye adsorption and cationic metal adsorption are favoured at pH > pHzpc, due to the presence of functional groups such OH^-, COO^- groups. Anionic dye adsorption and anionic metal adsorption are favoured at pH < pHzpc where the surface becomes positively charged [32,107]. The pHzpc values for some sawdust materials as examples are given in Table 3.

Zou et al. [32] observed that upon increasing the pH from 2 to 11, the amount of methylene blue adsorbed by *P. tabulaeformis* sawdust increased from 26.95 to 122.87 mg/g. This is due to the surface charge of the adsorbent, which becomes more negative at pH values well above the point of zero charge (pH_{PZC} 3.32). Adsorption was facilitated by electrostatic attraction. Below pH 3.32, the concentrations of H^+ ions were high and compete with the dye cation for adsorption sites. Similar tendencies were also noticed in the adsorption of dye ions from solutions by diverse sawdust materials [37,44,55,56,59,64,108].

Khattri and Singh [57] reported that the removal efficiency of crystal violet by *sagaun* sawdust increased with increasing pH values from 4 to 7.5; after pH 10, there was a decrease in dye ion removal. Similar trend was reported by Jain et al. [62]. The adsorption increased with the increase in pH from 3 to 6 and then decreased slightly in the range of pH between 7 and 9. Witek-Krowiak [63] also reported that uptake of malachite green by *beech* sawdust was minimal at pH 3 and increased with increasing pH from 7 to 12. Safiur Rahman and Rafiqul Islam [76] revealed that upon increasing pH from 2 to 8, the removal efficiency of Cu(II) by maple wood sawdust increased from 28.66 to 83.25%. At low pH, Cu(II) ions must compete with H^+ ions to attach the surface functional groups of the adsorbent. With the pH rise, sorption capacity increase due to the ionic charges on the metal and the ionic character of sawdust. Witek-Krowiak [77] explained the uptake of Cu(II) and Cr(III) on beech sawdust at pH 5 by attractive electrostatic interactions [77]. Gode et al. [109] found the opposite trends in Cr(VI) adsorption by modified red pine sawdust. Low pH values favour this system. Maximum adsorption occurred at pH 3. With increase in pH, the degree of protonation of carbon surfaces reduced and hence removal was decreased [109]. Similar tendencies were also noticed in the adsorption of chromium ions from solutions by diverse biosorbents [110].

3.2. Effect of initial concentration

Several studies that sought to clarify the effect of initial dye concentration on dye uptake by sawdust have been undertaken [12,19,32,42,44,49,54–60,62–64,111]. Ouazene and Sahmoune [42] reported that astrazon yellow uptake by aleppo pine-tree sawdust increased from 13.24 to 28.51 mg/g as the initial concentration of astrazon yellow increased from 15 to 50 mg/l. With increase in the initial dye concentration, the number of collisions between dye cations and sorbent increases, which enhances the sorption process. A similar trend was reported for the adsorption of dyes by diverse adsorbents [111]. The opposite trend can also exist [57,59,62]. Khattri and Singh [57] reported that the removal efficiency of crystal violet by *sagaun* sawdust decreased from 89.46 to 74.03% with an increase in initial crystal violet concentration from 6 to 12 mg/l.

Table 3
pHzpc values for different sawdust materials

Sawdust materials	pHzpc	Refs.
Beech sawdust	4.32	[77]
Citric acid-modified *pine* sawdust	3.32	[32]
Sawdust activated carbon (*Havea braziliansis*)	5.3	[106]
Maple sawdust	6.0	[107]
Meranti sawdust	6.23	[65]
Aleppo pine tree sawdust	6.8	[49]
Activated sawdust	7.5	[119]

3.3. Influence of temperature

It is well known that a temperature change alters the adsorption equilibrium in a specific way determined by the endothermic or exothermic nature of a process [112]. Zou et al. [32] reported that the uptake of methylene blue by *P. tabulaeformis* sawdust increased at higher temperature. The positive effect of temperature can be related to the enhancement of the diffusion rate of methylene blue through the boundary layer, and within internal pores of the sorbent particles [32]. The same behaviour can be detected in a research performed by Ahmad et al. [65,78] sequestering contaminant by sawdust material, suggesting that adsorption could take place by chemical ion exchange mechanism (endothermic process). These trends were also spotted by other research groups [38,41,78,79]. In contrast, Memon et al. [43] and Prado et al. [80] obtained the opposite behaviour for the temperature effect on adsorption capacity. Based on thermodynamic studies, Memon et al. [43] and Prado et al. [80] stated that Cd(II) and Pb(II) adsorption by sawdust materials are exothermic. A negative Gibbs free energy change (ΔG) indicates the feasibility and spontaneous nature of the adsorption process, whereas a negative enthalpy change (ΔH) represents the exothermic nature of an adsorption [43,80]. Djeribi and Hamdaoui [113] used *cedar* sawdust to adsorb copper(II). It was observed that with increase in temperature, adsorption capacity decreases indicating the adsorption is exothermic in nature. The negative values of (ΔG) and (ΔH) obtained from thermo dynamical analysis of the experimental data confirmed the spontaneous and exothermic nature of the adsorption. [113]. Ahmad [113] investigated the removal of trivalent chromium from its aqueous solution by spruce (*Picea smithiana*) sawdust by means of a radiotracer technique. Thermodynamic study showed spontaneous and exothermic nature of the sorption processes [114].

3.4. Effect of adsorbent dose

In principle, with more adsorbent present, the available adsorption sites or functional groups also increase. In turn, the amount of adsorbed dye ions or heavy metal ions is increased, which brought about improved adsorption efficiency [32,41,42,44,55,57,59,62,65,75,77–79].

Ouazene and Sahmoune [42] revealed that the removal efficiency of Astrazon Yellow by Aleppo pine tree sawdust increased from 30 to 91% with increasing dosage from 0.33 to 4 g/l and then remained almost constant. In contrast, Musapatika et al. [81] obtained the opposite behaviour on adsorption capacity of cobalt by pine sawdust. Authors stated that the decrease in adsorption capacity with increase in adsorbent dose could be due to the aggregation or overlapping of adsorption sites [81].

3.5. Effect of ionic strength

Miscellaneous inorganic chemicals, e.g. NaCl, KCl, $MgCl_2$, $CaCl_2$, etc. have been employed to elucidate the effect of ionic strength on dye or heavy metal uptake by sawdust material. As a general trend, the dye uptake or metal uptake is found to decrease with increasing ionic strength of the aqueous solution as a result of more electrostatic attraction and dye activity or metal activity change [32,36,43,109,115,116]. Zou et al. [32] observed that the effect of Ca^{2+} on the adsorption of the dye cations was more significant than that of Na^+ and K^+. The divalent Ca^{2+} ion had a greater affinity to the active sites and thereby might compete more strongly for binding sites than the monovalent Na^+ and K^+ ions. Djeribi and Hamdaoui [113] claimed that the adverse effect of ionic strength on copper uptake by *cedar* sawdust suggests the possibility of ion-exchange mechanisms being in operation in the sorption process and may be due to competition of Na^+ ions with copper cations for the same binding sites on the sorbent surface. Conversely, Memon et al. [43] explored that the presence of common anions such as acetate, bicarbonate, biphosphate, bromide, carbonate, citrate, sulphate and sulphide had no significant effects on the adsorption of Cd(II) by *Deodar cedrus* sawdust.

4. Desorption and regeneration of sawdust materials

Desorption studies help to elucidate the mechanism of adsorption and regeneration of adsorbent making the treatment process more economical and reasonable. Ansari et al. [44] reported that desorption data showed that nearly 99% of the congo red adsorbed on sawdust could be desorbed when ethanol was employed as eluting. Desorption studies were also carried out to elucidate the interactions between 2-picoline and the sawdust of walnut tree of Yazd [51]. Combustion was selected for 2-picoline desorption and the adsorbent was subsequently reused for more than three cycles. Functionalized sawdust has been evaluated for desorption of malachite green ions (MG) [59]. Desorption of MG was achieved by desorbing agents such as HCl, NaCl and NaOH using a batch reactor system. Both these desorbents were suitable to remove the adsorbed MG from the biomass. It was found that HCl 1 N could desorb 92.28% of MG

initially loaded on to the biomass. Treated sawdust has been evaluated for desorption of methylene blue (MB). [67]. It was found that HCl could desorb more than 90% of MB initially loaded on to the biomass. Khattri and Singh [68] reported desorption of MG laden neem sawdust. 46% desorption of MG from Neem sawdust was achieved using 1.5% KCl solution. Desorption studies were also performed by Ofomaja and Ho [72] and more of the adsorbed methyl violet was desorbed at lower pH. Desorption was reduced as solution pH increased to pH 7. Witek-Krowiak [77] studied desorption of Cu(II) and Cr(III) from previously loaded by *beech* sawdust using 0.1 M HCl. Desorption data showed that after four cycles, the decrease in the efficiency did not exceed 30%. Sidiras et al. [82] reported that pure water desorbed Cr(VI) from both untreated and pretreated *pine* sawdust. The results were 23.3–48.8% Cr(VI) desorption for untreated *pine* sawdust and 5.8–17.6% Cr(VI) desorption for pretreated pine sawdust. Studies were also carried out on the regeneration of sawdust by Larous et al. [115]. The desorption percentage of copper on sawdust was 33.8% for NaCl. The other extractants were not efficient in desorption of Cu(II) ions. Desorption of Cr(VI) from sawdust using acid and base treatment exhibits higher desorption efficiency by more than 95% [117]. In contrast, Gode et al. [109] found that desorption of Cr(VI) from modified red *pine* sawdust using acidic pH exhibited lower desorption efficiency.

5. Conclusions and future perspectives

From the above review, it can be observed that a number of sawdust materials has been tested as an adsorbent for the removal of dyes and heavy metals in batch mode experiment.

There are very limited column studies and no report on commercial plant.

Although advances have been accomplished in terms of sorption properties and kinetics, much work remains undone and is necessary to identify clearly the sorption mechanisms.

On the basis of the results obtained from the IR analysis and pH, kinetic, isotherm studies and desorption, we conclude that the adsorption mechanism, underlying the sorption of dyes and heavy metals onto sawdust materials might be partly ion exchange, chemisorption, electrostatic interaction and intraparticle diffusion.

In order to fully utilize the application of these sawdust materials in wastewater treatment, their removal efficiency should be further tested in effluents from industries. Pilot-plant scale studies and cost evaluation remain to be explored. As a general trend, studies on the adsorption of dyes and heavy metals by sawdust revealed that Langmuir and pseudo-second-order models were best fitted to the experimental data. Of process factors, pH appears to be the most influential. Sawdust is effective in heavy metal and dye uptake especially after chemical treatments. Sawdust can be used as an adsorbent instead of activated carbon and replace the regeneration step by making use of composting the adsorbent. Composting the spent adsorbent will degrade the adsorbent material. The degraded product is a stabilized product which is added to soil to improve soil structure, especially for clay soils, or which acts as a fertilizer improving the nutrient content.

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